Pressure effect on superconductivity of A_xFe$_2$Se$_2$ (A = K and Cs)

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We performed the high hydrostatic pressure resistivity measurements (up to 1.7 GPa) on the newly discovered superconductors A_xFe$_2$Se$_2$ (A = K and Cs) single crystals. Two batches of single crystals $K_xFe_2Se_2$ with different transition temperatures (T_c) were used to study the effect of pressure. The T_c of the first one gradually decreases with increasing pressure from 32.6 K at ambient pressure. While a dome-like behavior was observed for the crystal with T_c = 31.1 K, and T_c reaches its maximum value of 32.7 K at the pressure of 0.48 GPa. It indicates that there exists a optimal doping with maximum T_c of 32.7K in $K_xFe_2Se_2$ system. The behavior of T_c vs. pressure for $Cs_xFe_2Se_2$ also shows a dome-like behavior, and T_c reaches its maximum value of 31.1 K at the pressure of 0.82 GPa. The hump observed in temperature dependence of resistivity for all the samples tends to shift to high temperature with increasing pressure. The resistivity hump could arise from the vacancy of Fe or Se.

The newly discovered iron-based superconductors have attracted much attention in past three years[1–5]. Up to now, various Fe-based superconductors, such as ZrCuSiAs-type LnFeAsO (Ln is rare earth elements)[1–3], ThCr$_2$Si$_2$-type AeFe$_2$As$_2$ (Ae is alkali earth elements)[5], Fe$_2$As-type AFeAs (A is Li or Na)[6–8] and anti-PbO-type Fe(Se,Te)[9], have been reported. The T_c of anti-PbO-type FeSe could reach 37 K under 4.5 GPa from T_c∼8K at the ambient pressure[10]. Very recently, by intercalating K, Rb, Cs and Tl into between the FeSe layers, superconductivity has been enhanced to around 30 K without any external pressure in Fe-Se system[11–16], it provide a new type of iron-based superconductor to explore high T_c. For the iron-pnictide, the pressure tends to destroy the magnetic transition in the undoped compounds, and T_c increases with increasing pressure for underdoped iron-pnictides, and remains approximately constant for optimal doping, and decreases linearly in the overdoped range[17,18]. Superconductivity can be induced by pressure in the parent compounds AFe_2As_2 (A=Ca, Sr, Ba, Eu)[19–21]. Magnetism and superconductivity are strongly correlated with each other in the iron-based superconductors. The strong pressure effect in FeSe may be related to its strongly enhanced antiferromagnetic spin fluctuations under pressure[22]. Therefore, we wonder weather the strong pressure effect still exist in $A_xFe_2Se_2$. It is very meaningful to perform high pressure measurement in this newly found superconductors.

In this paper, we systematically measured resistivity under the high hydrostatic pressure up to 1.7 GPa for the newly discovered superconductors $K_xFe_2Se_2$ and $Cs_xFe_2Se_2$. It is found that the transition temperature slightly increases below 0.82 GPa, and gradually decreases with further increasing the pressure for $Cs_xFe_2Se_2$. Two $K_xFe_2Se_2$ single crystals with different T_c were measured. For the $K_xFe_2Se_2$ crystal with T_c^{onset}=32.6 K and broad hump centered at 245 K, and T_c gradually decreases with increasing pressure. While T_c vs. pressure shows the similar behavior to $Cs_xFe_2Se_2$, and T_c^{onset} reaches its maximum value 32.7 K under the pressure of 0.48 Gpa for the $K_xFe_2Se_2$ with T_c^{onset}=31.1 K and broad hump centered at 130 K. All these results suggest that there exists an optimal doping with maximum T_c. T_c increases with increasing pressure for underdoped sample, and monotonically decreases in the overdoped range. Resistivity hump shifts to high temperature with increasing the pressure.

Single crystals $A_xFe_2Se_2$(A=K,Cs) was grown by self-flux method as described elsewhere.[14] Many shinning plate-like single crystals can be cleaved from the final products. $Cs_xFe_2Se_2$ shows superconductivity at about 30 K, the actual compositions determined by EDX is $Cs_{0.86}Fe_{1.66}Se_2$. Two different types of $K_xFe_2Se_2$ were obtained. The actually composition of the first one with the T_c^{onset}=32.6K is $K_{0.85}Fe_2Se_{1.80}$(denoted $K_xFe_2Se_2$-1). The actually composition of the second one with the T_c^{onset}=31.1K is $K_{0.86}Fe_2Se_{1.82}$(denoted $K_xFe_2Se_2$-2). Pressure was generated in a Teflon cup filled with Daphne Oil 7373 which was inserted into a Be-Cu pressure cell. The pressure was determined at low temperature by monitoring the shift in the superconducting transition temperature of pure tin. The measurement of resistivity was performed using the *Quantum Design* PPMS-9.

Fig.1(a), (b) and (c) shows the temperature dependence of the in-plane resistivity for single crystals $K_xFe_2Se_2$-1, $K_xFe_2Se_2$-2 and $Cs_xFe_2Se_2$ under different pressures. $K_xFe_2Se_2$-1 shows the semiconducting behavior at the high temperature and displays a broad hump at about 245 K under ambient pressure, and shows superconductivity at 32.6 K. The resistivity gradually decreases and the hump becomes much more unobvious with increasing the pressure. For $K_xFe_2Se_2$-

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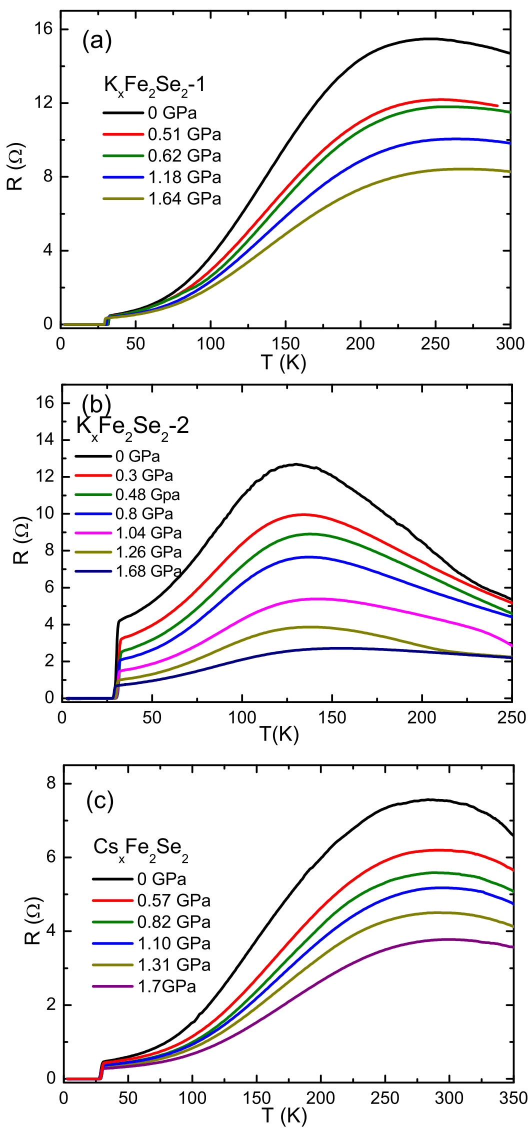

FIG. 1: (Color online) Temperature dependence of the in-plane resistivity under different pressures for the single crystals: (a). $K_xFe2Se2$-1; (b), $K_xFe2Se2$-2; and (c), $Cs_xFe2Se2$.

2, the broad hump occurs at 130 K and T_c^{onset} is 31.1 K. For $Cs_xFe_2Se_2$, similar resistivity behavior was observed with the broad hump centered at around 285K. With increasing the pressure, resistivity remarkably decreases and the hump feature becomes much more obscured.

Fig.2(a) shows the temperature dependence of the resistivity of $Cs_xFe_2Se_2$ under the different pressures in the low temperature range. It indicates that the resistivity gradually decreases with increasing pressure. We defined the T_c with the temperature at which the resistivity drops 90%, 50% and 10% relative to the resistivity just above the superconducting transition. Fig.2(b)

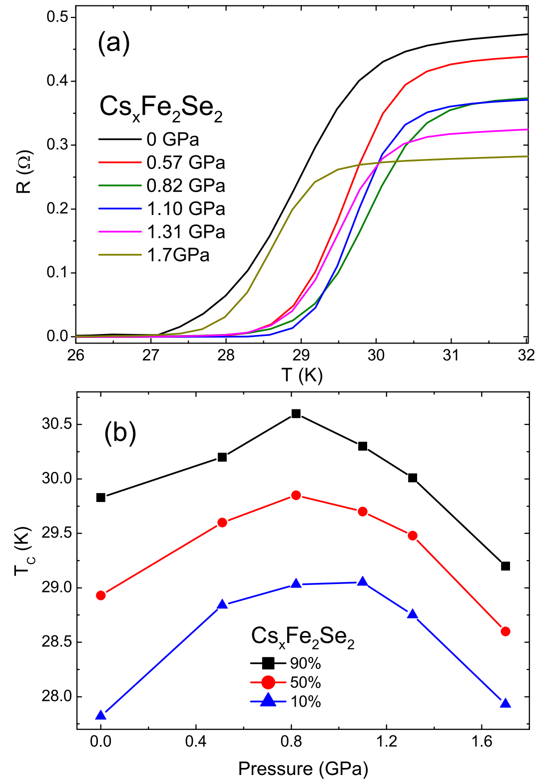

FIG. 2: (Color online) (a): Temperature dependence of resistivity for single crystal $Cs_xFe_2Se_2$ under the different pressures around superconducting transition temperature range; (b): Pressure dependence of T_c for single crystal $Cs_xFe_2Se_2$.

shows the pressure dependence of T_c. T_c increases with increasing the pressure below 0.82 GPa, and T_c gradually decreases with further increases the pressure. T_c^{onset} increases to 31.1K under the pressure of 0.82 GPa from the 30K at ambient pressure. dT_c/dP∼1.3K/GPa in the region of P<0.82 GPa is much less than that in FeSe, and almost the same as that observed in the electron-doped LaOFeAs[24]. The pressure dependence of T_c in $Cs_xFe_2Se_2$ is quite similar to that of $LaO_{1-x}F_xFeAs$ system[25].

Fig.3(a) shows the temperature dependence of the resistivity of $K_xFe_2Se_2$-1 under the different pressures in the low temperature range. The resistivity gradually reduced with increasing pressure in the normal state. Fig.3(b) shows the pressure dependence of T_c. T_c monotonically decreases with increasing pressure, being different from that shown in Fig.2b for the $Cs_xFe_2Se_2$. T_c^{onset} decreases to 29.8 K at the pressure of 1.64 GPa, which is about 2.8 K lower than the T_c at ambient pressure. The behavior of T_c vs. pressure for this compound is consistent with the previous report[23], while quite different from the report by Kawasaki et al.[26] Kawasaki et al. reported that T_c^{onset} increases with increasing pres-

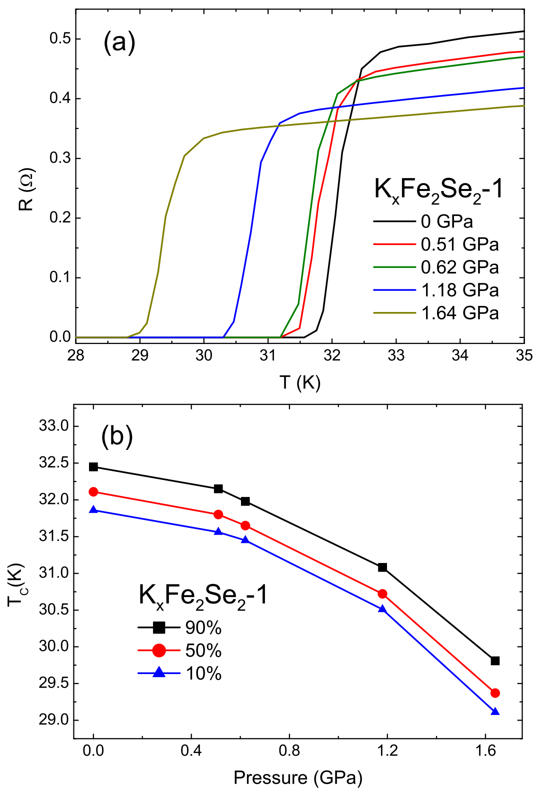

FIG. 3: (Color online) (a): Temperature dependence of resistivity for single crystal $K_xFe_2Se_2$-1 under the different pressures around superconducting transition temperature range; (b): Pressure dependence of T_c for single crystal $K_xFe_2Se_2$-1.

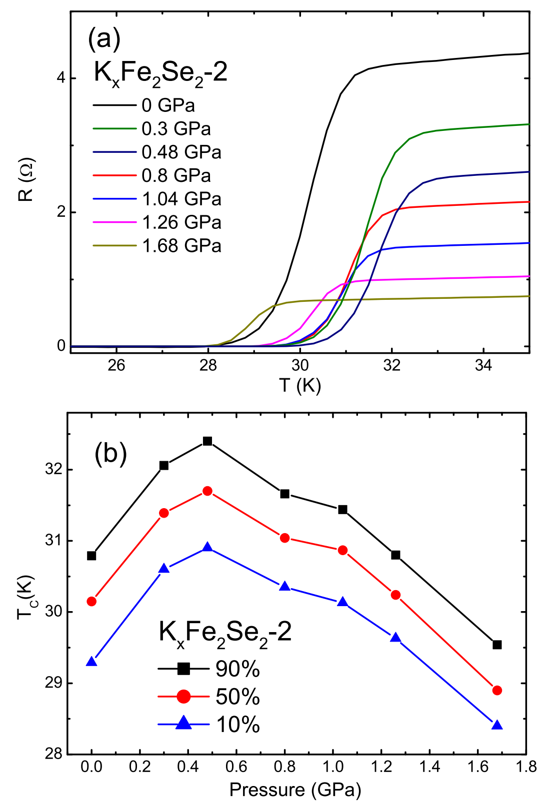

FIG. 4: (Color online) (a): Temperature dependence of resistivity for single crystal $K_xFe_2Se_2$-2 under the different pressures around superconducting transition temperature range; (b): Pressure dependence of T_c for single crystal $K_xFe_2Se_2$-2.

sure, while T_c^{zero} decreases. It suggests that the superconducting transition becomes broadening with increasing pressure. However, the superconducting transition width nearly does not change by pressure as shown in Fig.3(a). Such difference could arise from the quality of single crystal or inhomogeneity of applied pressure.

Fig.4(a) shows the temperature dependence of the resistivity of $K_xFe_2Se_2$-2 under the different pressure around the temperature range of superconducting transition. The T_c at ambient pressure is 1.5 K lower than that of $K_xFe_2Se_2$-1. The behavior of T_c vs. pressure is quite different from that of $K_xFe_2Se_2$-1 as shown in Fig.3(b). T_c as a function of pressure shows a dome-like behavior as shown in Fig.4(b). It is similar to that observed in single crystal $Cs_xFe_2Se_2$. T_c^{onset} gradually increases with increasing pressure and reaches the maximum value of 32.7 K at the pressure of 0.48 GPa, then monotonically decreases with further increasing pressure. It should be addressed that the $K_xFe_2Se_2$-1 and $K_xFe_2Se_2$-2 shows different pressure dependence of T_c. Such different pressure dependence of T_c indicates that there exists optimal doping with the maximum $T_c \sim 32.7$ K in $K_xFe_2Se_2$ system. The pressure tends to destroy the magnetic transition in the undoped compounds and T_c increases with increasing pressure for underdoped iron-pnictides, and monotonically decreases in the overdoped range. The different effect of pressure on T_c between the crystals $K_xFe_2Se_2$-1 and $K_xFe_2Se_2$-2 is easily understood because the $K_xFe_2Se_2$-1 is in the slightly overdoped range, while the $K_xFe_2Se_2$-2 is in the underdoped range.

Fig.5 shows the pressure dependence of the resistivity hump temperature for $Cs_xFe_2Se_2$, $K_xFe_2Se_2$-1 and $K_xFe_2Se_2$-2. We defined the hump temperature when the resistivity reaches its maximum value. The temperature of the hump monotonically increases with increasing pressure for all the samples. The shift of the resistivity hump temperature for $Cs_xFe_2Se_2$ is very small below 0.82 GPa and increases apparently in the high pressure range. The temperature of the hump shows no direct connection with the T_c because the pressure dependence of hump temperature is quite different from that of T_c. The origin of the hump in this kind of superconductors could arise from the content of K or the vacancy of Fe or Se sites.

The normal state resistivity behavior and pressure dependence of T_c are quite different for the crystals

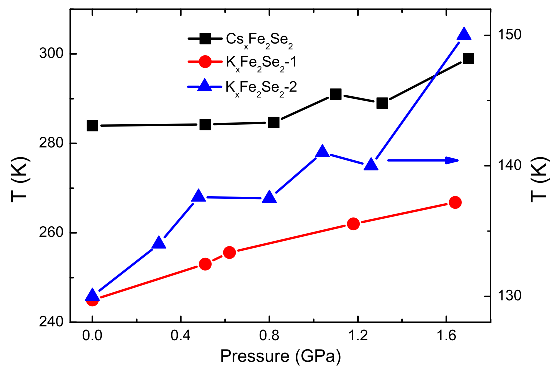

FIG. 5: (Color online) Pressure dependence of the resistivity hump temperature for single crystals $Cs_xFe_2Se_2$, $K_xFe_2Se_2$-1 and $K_xFe_2Se_2$-2.

$K_xFe_2Se_2$ with different T_c. The T_c as a function of pressure for $K_xFe_2Se_2$-2 shows a dome-like behavior and reaches its maximum T_c at the pressure of 0.48 GPa. While T_c is gradually suppressed with increasing the pressure for $K_xFe_2Se_2$-1. The pressure dependence of T_c for $K_xFe_2Se_2$-2 and $Cs_xFe_2Se_2$ is quite similar to that observed in the $LaO_{1-x}F_xFeAs$ and parent iron-pnictides[21,25,27]. While for $K_xFe_2Se_2$-1, the pressure dependence of T_c is similar to that observed in the over-doped iron-pnictides[28]. The maximum T_c for the two batched of single crystals $K_xFe_2Se_2$ is the same, and reaches at ambient pressure and at 0.48 Gpa, respectively. The difference of T_c between the two batches of single crystals $K_xFe_2Se_2$ is very small (just 1.5 K), while the temperature of the resistivity hump is 245 K and 130 K, respectively. It indicates the hump temperature strongly depends on the sample. The hump behavior could originate from the vacancy of Fe or Se. Another evidence is that the normal state resistivity is very high compared with other iron-pnictide superconductors because the vacancy in conducting FeSe layers leads to a strong scattering, consequently high resistivity. It suggests that the physical behavior of $A_xFe_2Se_2$ is very sensitive to the deficiency, and change of the deficiency strongly affects the normal state resistivity although the T_c does not change too much.

In conclusion, we performed the high hydrostatic pressure resistivity measurement for the newly discovered superconductors $A_xFe_2Se_2$(A=K, Cs). For $Cs_xFe_2Se_2$, T_c starts to increase at the pressure less than 0.82 GPa, and T_c decreases with further increasing the pressure. This behavior is similar to that in $K_xFe_2Se_2$-2. While the behavior is quite different for $K_xFe_2Se_2$-1 with T_C^{onset}=32.6K, T_c monotonically decreases with increasing pressure. The different pressure dependence of T_c between these single crystals is because they are in different doping level with different T_c. The temperature of resistivity hump increases with increasing the pressure. The resistivity hump could arise from the deficiency of Fe or Se in conducting layers.

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